Radioactive Isotopes of Sr, Y and Zr

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Radioactive isotopes formed by bombardment of Rb, Sr and Y by protons, deuterons and neutrons are reported. The following are the periods and assignments: 2.75-hr. (Sr^{87*}, e^-, γ); 70-min. (Sr^{85}, e^-, γ); 66-day (Sr^{85}, K, γ); 80-hr. (Y^{87}, K); 14-hr. (Y^{87}, e^-, γ); 105-day (Y^{86}, K, γ); 4.5-min. (Zr^{89}, γ); 78-hr. (Zr^{89}, β^+). (In each case e^- means conversion electrons, not nuclear β-rays.) The electron spectrum of the 2.75-hr. Sr^{87*} shows a single line at 360 kev and this period is shown to grow from the 80-hr. Y^{87}, but not from the 14-hr. isomer.

The bombardment of Rb, Sr and Y targets by 6.7-Mev protons, 4.5-Mev deuterons and slow and fast neutrons produces at least twelve radioactive periods. We have studied ten of these, many of which have also been studied by Stewart, Lawson and Cork[1] and by Stewart.[2] Our results show several periods not reported by them and an important change of assignment of one Sr period.[3] The stable isotopes of Sr, Y and Zr together with the probable assignments of the radioactive periods are shown in Fig. 1.[4]

The targets were usually in the form of chloride salts or in the case of Sr, the pure element. $SrCl_2$ could also be satisfactorily bombarded when fused onto a stainless steel plate contained in an evacuated cup attached to the exit window of the cyclotron. Beam currents to the cup were usually of the order of 1 or 2μa of protons or deuterons. In some cases targets were bombarded on an internal probe where the beam current is about 20μa. Activities were followed with a Freon-filled ionization chamber and d.c. amplifier.[5] A magnetic cloud chamber and a β-ray spectrograph were used to study β-ray and γ-ray spectra while a Geiger-Müller counter with a scale-of-eight was sometimes used for absorption measurements.

To separate Sr chemically from Rb the RbCl was dissolved in water and Na_2CO_3 was added in excess. There was no attempt made to recover the rubidium. Yttrium was separated from Sr either as the hydroxide by the addition of NH_4OH or, in the later experiments, with 8-hydroxyquinoline (oxine) by using a technique originally developed for the separation of Al from alkaline earths.[6] The method is as follows: 0.17 g of oxine is dissolved in 12.5 cm^3 of acetone and the solution is diluted to about 50 cm^3 with water. About 5 mg of YCl_3 is added as a carrier to the bombarded Sr which is dissolved in 15 cm^3 of water. 7.5 cm^3 of the oxine solution is added to this solution and NH_4OH is added by drops until the ammonia odor is noticeable. This solution is warmed on a water bath to coagulate the precipitate and is filtered out in a Gooch crucible containing filter paper. This method has the advantage that it gives more nearly complete separations than the hydroxide method and also gives a crystalline precipitate which is more rapidly filterable.

Zirconium was separated from bombarded Y as the iodate in nitric acid solution, with a trace of $Zr(NO_3)_4$ added as carrier.

In all cases the filtered precipitate was left on a disk of filter paper 2 cm in diameter and

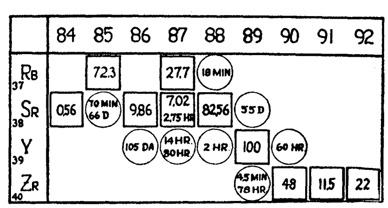

Fig. 1. Stable isotopes with relative abundance and probable assignment of radioactive periods.

[1] Stewart, Lawson and Cork, Phys. Rev. **52**, 901 (1937).
[2] D. W. Stewart, Phys. Rev. **56**, 629 (1939).
[3] L. A. DuBridge and J. Marshall, Phys. Rev. **56**, 706 (1939); **57**, 348A (1940).
[4] Our results, together with other unpublished data from this laboratory, were transmitted to Dr. G. T. Seaborg for inclusion in the table by J. J. Livingood and G. T. Seaborg, Rev. Mod. Phys. **12**, 30 (1940).
[5] S. W. Barnes, Rev. Sci. Inst. **10**, 1 (1939).
[6] F. L. Hahn and K. Vieweg, Zeits. f. anal. Chemie **71**, 122 (1927).

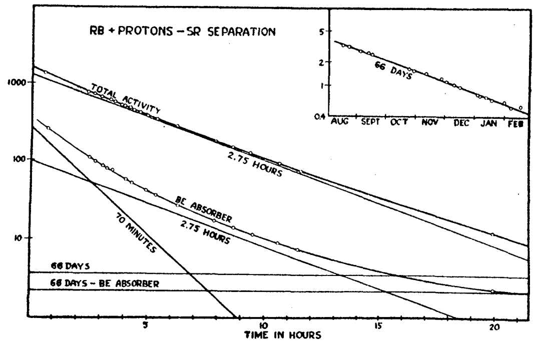

FIG. 2. Decay of Sr from Rb+H^1. Upper curve, no absorbers, lower curve, 2 mm of Be.

held in place with Scotch tape, which provided a convenient form for the measurement of radioactivity.

1. Radioactive Isotopes of Sr

Bombardment of Rb by protons produces a strong activity in the Sr fraction whose decay curve is shown in Fig. 2. The chief component is a period of 2.75 ± 0.1 hr. If the bombarded Rb sample is covered with a Be plate 2 mm thick the 2.75-hr. period is greatly reduced in intensity while a period of 70 minutes and one of 66 days become evident (see lower curve). The 70-min. period is almost completely masked without the Be absorber. This Be plate has an absorbing power equal to about 90 mils of Al for β-rays but has negligible absorption for x-rays and γ-rays. Apparently therefore very few charged particles are associated with these 70-min. and 66-day periods, the radiation consisting largely of x-rays and γ-rays. The 2.75-hr. activity consists largely of negative electrons.

There are only two stable isotopes of Rb, of masses 85 and 87, the latter being naturally β-active with a period of 10^{10} years. Rb(p,n) reactions therefore would yield only Sr^{85} and Sr^{87}. Since Sr^{87} is stable it would appear necessary at first sight to assign all three periods to Sr^{85}. However it will be shown below that the 2.75-hr. period must be assigned to a metastable state of Sr^{87}. The 70-min. and 66-day periods are therefore assigned to Sr^{85}. Since Rb^{84} does not exist and Sr^{84} is very rare these Sr^{85} periods are not produced in observable amounts by deuteron or neutron bombardment of Rb or Sr.

Sr^{85} (70 min. and 66 day)

As shown in Fig. 2 the activity of these two periods is only slightly reduced by covering the sample with Be. Since no positrons can be observed and since Y^{85} is not stable it is concluded that this isotope decays by K capture to Rb^{85}. The γ-ray spectrum of the short period can not easily be obtained because of the presence of the strong 2.75-hr. period. However in β-ray spectrograph plates taken to get the electron spectrum of this latter period, a faint electron line is observed at 160 kev. (Fig. 3.) Since this line does not appear when the 2.75-hr. period is formed in other ways it can probably be associated with the 70-min. Sr^{85}. It seems probable then that the 70-min. isomer decays to the 66-day isomer with the emission of a partly converted γ-ray of about 170 kev (the K-ionization energy for Sr being about 10 kev).

The decay of the 66-day isomer by K capture to Rb^{85} is accompanied by a γ-ray whose absorption coefficient in lead indicates an energy of about 0.8 Mev.

Sr^{87*} (2.75 hr.)

As indicated in an earlier communication[3] this period is the same as the 3.0-hr. period reported by Stewart Lawson and Cork[1] and by Stewart.[2] It was formed in their experiments by Sr+d and Sr+n and assigned to Sr^{89}, isomeric with the 55-day period. The formation of this activity by protons on Rb eliminates this possible assignment. Its formation by Sr+d and Sr+n would allow its assignment to Sr isotopes 85 (weak), and 87 as well as 89. Since Sr^{87} is stable it would be natural to make the assignment to Sr^{85}. However it is found that the same period is also formed by Sr+p. The only way in which a Sr activity could be formed from Sr+p is either by a $p-p$ reaction leading to a metastable state of a stable isotope, or by decay of an yttrium activity formed during the bombardment. If the latter is formed by a $p-n$ process the decay product must also be an isomeric state of a stable nucleus thus eliminating assignment to Sr^{85}. As will be seen below the 2.75-hr. period does indeed grow from an 80-hr. Y activity formed both by Sr+p and Sr+d, an activity which can be independently assigned to Y^{87}.

This makes the assignment of the 2.75-hr. period to a metastable state of Sr^{87} quite certain.

In the $Sr+n$ bombardments an activity assigned to Sr^{87*} could be formed either by the $n-\gamma$ or the $n-n$ excitation process. We have mounted a pure Sr sample in a Cd box immediately behind a Be probe within the cyclotron chamber. Since it was there exposed to a high intensity of medium fast neutrons and but few slow neutrons the $n-\gamma$ process would probably be unimportant. On the other hand, the neutrons formed by $Be+p$ are not fast enough to cause an $n-2n$ process. The 2.75-hr. period was produced under these conditions with fair intensity, suggesting the $n-n$ process. By mounting an In sample of similar dimensions adjacent to the Sr sample an estimate of the relative yields of the two reactions $Sr^{87}(n,n)Sr^{87*}$ and $In^{115}(n,n)In^{115*}$ (4.1 hr.)[7] could be made. The yield for the latter process (calculated for infinite bombardment of the pure isotope) is about 2 to 4 times as great as for the former. The actual ratio may be greater than this since the $n-\gamma$ process may contribute in the first case (Sr^{86} being fairly abundant) but not in the second (since In^{114} is not stable). This difference in yields may be connected with the fact that the spin of the ground state of In^{115} is 9/2 and of Sr^{87} is $\frac{1}{2}$. Hence the spin of the isomeric In^{115} is probably $\frac{1}{2}$ and of Sr^{87*} probably 9/2.

The electron and γ-ray spectrum of this 2.75-hr. period is of particular interest. If it is assigned to either Sr^{85} or Sr^{87*} the decay must be either by positron emission or K capture since the corresponding Y isotopes are not stable. The cloud-chamber measurements of Stewart *et al.*[1,2] showed an apparently continuous

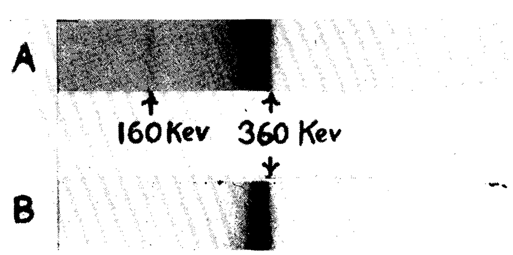

FIG. 3. (A) Electron spectrum of Sr from $Rb+H^1$. (70 min., 2.75 hr.) (B) Spectrum of Sr bombarded with protons. (80-hr. period).

[7] Goldhaber, Hill and Szilard, Phys. Rev. **55**, 47 (1939).

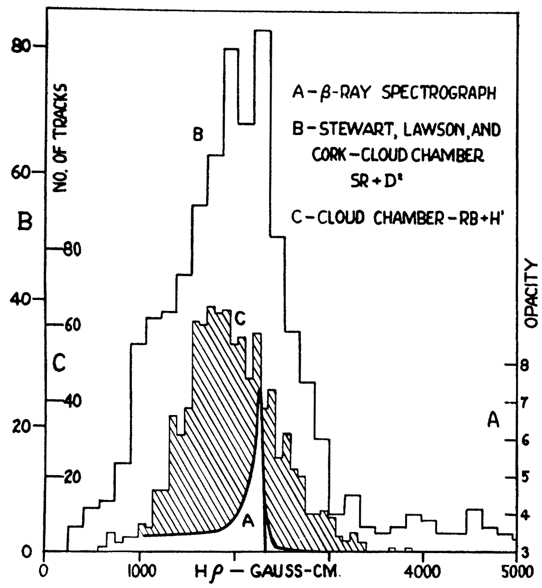

FIG. 4. Comparison of cloud chamber and β-ray spectrograph measurements of the electron spectrum of 2.75-hr. Sr.

spectrum of low energy negative electrons. This was indeed a strong reason for assigning the period to Sr^{89} where it could decay by electron emission to stable Y^{89}. Our cloud-chamber histogram (C) is compared to theirs (B) in Fig. 4. For clarity the two are drawn to different scales. The high energy tracks above $H\rho$ 3300 in curve B are from the 55-day Sr^{89} also present in their specimen, but not in ours. Stewart, Lawson and Cork found this histogram to fit a K$-$U plot.

However, when a thin specimen showing this activity is placed in the β-ray spectrograph a spectrum consisting only of a single line at $H\rho$ 2360 is obtained. Two such plates are shown in Fig. 3, one taken with a $Rb+p$ specimen and the other from $Sr+p$. Exactly similar plates were obtained from a $Sr+d$ bombardment. The microphotometer curve for one of these plates is plotted as A in Fig. 4, showing a much narrower peak than the cloud-chamber data. It is evident that for electrons of this low energy (360 kev) the scattering of the particles in the cloud-chamber gas (plus self-absorption in the specimen if thick) broadens out the histogram to such an extent as to conceal the homogeneity of the electron spectrum.

The results with the β-ray spectrograph make it certain that the observed electrons are con-

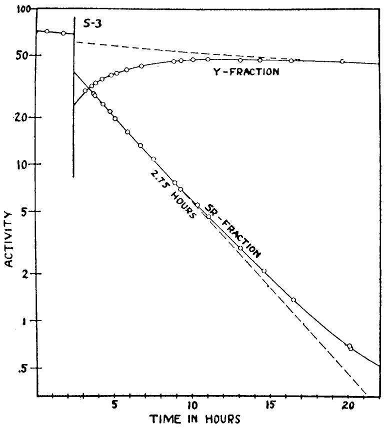

FIG. 5. Typical curves of the growth and decay of activity in Y and Sr fractions from previous Y separation from $Sr+H^1$.

version electrons accompanying a transition gamma-ray and are not nuclear β-rays. The final necessity of assignment of this period to Sr^{89} is removed and the assignment to Sr^{87*} becomes unambiguous.

That Sr^{87*} decays by isomeric transition to the stable state Sr^{87} rather than by K capture to Rb^{87} is almost certain since Rb^{87} is itself naturally β-active decaying to Sr^{87}. This latter decay must always be to the ground state of Sr^{87} since the maximum β-ray energy is[8] only 130 kev and there is no evidence in the Rb^{87} spectrum of the Sr^{87*} electrons. Also we have been unsuccessful in separating active Sr from natural Rb.[3]

That the x-rays accompanying the Sr^{87*} transition are Sr rather than Rb K x-rays was shown roughly by absorption experiments in Se and Br. Thin evaporated layers of Se on Al foil and distilled layers of CBr_4 on Al were used as absorbers. The absorption coefficient in Br was of the same order of magnitude as in Se, whereas for Rb K x-rays it should have been several times smaller. The measurements are only rough because the only samples of the 2.7-hr. period strong enough for these tests were those formed by Rb(p,n) in which the Sr^{85} periods were also strong.

2. Radioactive Isotopes of Y

Bombardment of Sr with protons yields four periods in Y. A 2-hr. positron emitting period has been observed by Stewart, Lawson and Cork.[1] They assign it to Y^{88} formed by $Sr^{87}(d,n)$ and $Y^{89}(n,2n)$. We have evidence that it is also formed by $Sr^{88}(p,n)$, but have no good determination of the period because of the masking effect of Sr^{87*}.

In addition to the 2-hr. period we have observed three other periods produced by protons: 14 ± 2 hours, 80 ± 3 hours, and 105 ± 5 days. Since the 14-hr. and the 80-hr. period are also produced by the bombardment of Sr with deuterons (also observed by Stewart) they can be assigned only to Y^{87} or Y^{88}. Stewart, Lawson and Cork, however, found only the 2-hr. period as a result of the reaction $Y^{89}(n,2n)$. Therefore, we must assume that the 14-hr. and 80-hr. period are isomers of Y^{87}.

Beta-ray spectrograph measurements showed that the electrons which accompany the 80-hr. period have a line spectrum identical with that of the 2.7-hr. Sr, which period was then not yet definitely assigned. When an aged Y fraction from a $Sr+p$ or $Sr+d$ target is dissolved and Sr and Y separations again made, the Sr fraction decays with the 2.7-hr. period and the Y fraction activity grows with this period. A typical pair of curves is shown in Fig. 5. This shows that the 2.7-hr. period grows from the decay of the 80-hr. period confirming the assignment to Sr^{87*} and Y^{87}, respectively.

The decay curves for five such successive separations are shown in Fig. 6, correction being made for the loss (of about 10 percent) in each separation. The initial activities of the successive Sr fractions fall off with the 80-hr. period,

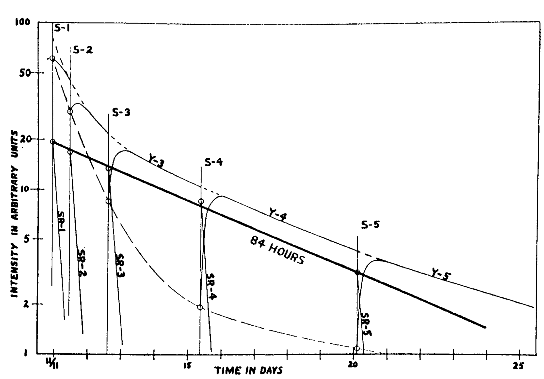

FIG. 6. Multiple separations from $Sr+H^1$. Normalized at each separation to compensate for loss in chemical separation.

[8] W. F. Libby and D. D. Lee, Phys. Rev. 55, 245 (1939).

showing that the isomeric 14-hr. period of Y^{87} does not decay to Sr^{87*}. Presumably this 14-hr. state decays to the 80-hr. state with gamma-ray emission.

Also the initial activities of the Y fractions decay along a curve which does not show any 80-hr. component, indicating that practically the entire activity of this period is assignable to the Sr^{87*} conversion electrons. When these electrons are filtered out with a Be plate there is, however, evidence for the $Sr-K$ x-rays which accompany the K-capture decay of Y^{87} to Sr^{87*}. We have detected no 80-hr. gamma-ray, however.

The 14-hr. isomeric transition in Y^{87} appears from absorption measurements (made immediately after separation) to be accompanied by a gamma-ray of about 500 kev and a few corresponding conversion electrons. Measurements of these are made difficult, however, by the rapid rise of the 2.7-hr. activity.

The 100-day period appears to decay by K-electron capture and the emission of a fairly high energy unconverted γ-ray. Absorption measurements of the γ-ray in Pb, Al, and Cu indicate a γ-ray of about 2 Mev, but absorption measurements at these energies are quite uncertain and the γ-ray may even be as low as 1 Mev in energy, or may be a mixture of two or more. The fact that the 100-day period is formed by protons on Sr, but not by deuterons makes the most probable assignment Y^{86}.

In one of the earlier multiple separations after deuteron bombardment of Sr we found a 15 ± 2-day period in the Y fraction. We assigned it tentatively to Y^{85} since it was not observed as a product of proton bombardment. This separation was done by the hydroxide method. A later attempt to find this period was unsuccessful with the oxine method of separation, and hence was undoubtedly due to an impurity (P^{32}?).

TABLE I. *Characteristics of radioactive isotopes of* Sr, Y *and* Zr. *I.T.=isomeric transition,* e^-*=conversion electrons,* β^+*=positrons,* $K=K$ *capture.*

PERIOD	EMITTED PARTICLE	ASSIGNMENT	FORMED BY	ENERGY (MEV)
2.75 ±0.1 hr.	e^-, γ I.T.	Sr^{87*}	Rb-p-n, Sr-d-p Sr-n-γ, Sr-n-n Sr-p-p(?), $Y^{87}+e_K$	e^-, 0.360
70 min.	I.T., e^-, γ	Sr^{85}	Rb-p-n	e^-, 0.160(?)
66 day	K, γ	Sr^{85}	Rb-p-n	γ, 0.8
80 hr.	K (to Sr^{87*})	Y^{87}	Sr-p-n, Sr-d-n	no γ(?)
14 hr.	I.T., e^-, γ	Y^{87}	Sr-p-n, Sr-d-n	e^-, γ, $\sim$0.5
105 day	K, γ	Y^{86}	Sr-p-n	$\gamma\sim$2.0(?)
4.5 min.	I.T., γ	Zr^{89}	Y-p-n	
78 hr.	β^+	Zr^{89}	Y-p-n	$\beta^+\sim$1.0 no γ

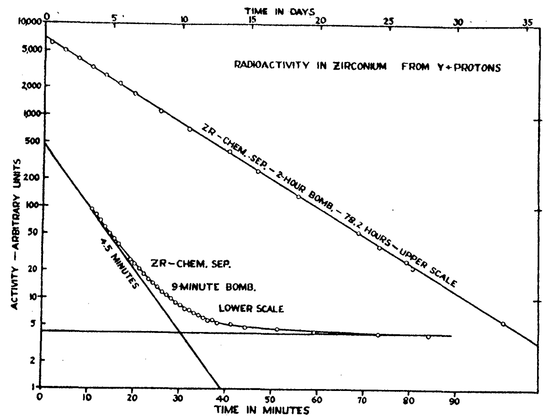

FIG. 7. Decay of Zr from $Y+H^1$.

3. Zr PERIODS

The bombardment of Y with protons produced two strong periods of 4.5 min. and 78 ± 1 hour (see Fig. 7), which are chemically identified as Zr. The 4.5-min. period appears to be associated with an isomeric transition in Zr^{89}. This is shown by the cloud-chamber observation that many γ-rays are present while the 4.5-min. period is strong, but after this period has died out only positrons are observable. Range measurements in Al show these to have a maximum energy of about 1 Mev. There are no γ-rays associated with the 78-hr. period.

These two activities must be isomers of Zr^{89} because no other Zr isotope can be produced from Y by a $p-n$ reaction.

The results reported herein are summarized in Table I. The 55-day Sr^{89} and the 2-hr. Y^{88} are omitted since though we observed them we have nothing to add to the data of Stewart.[2]

The authors are indebted to many members of the laboratory for help with this work. Especially we wish to thank Mr. R. L. McCreary and Dr. G. E. Valley for assistance with the cloud chamber and the β-ray spectrograph, and Mr. Gerhard Dessauer for assistance with the chemical separations.

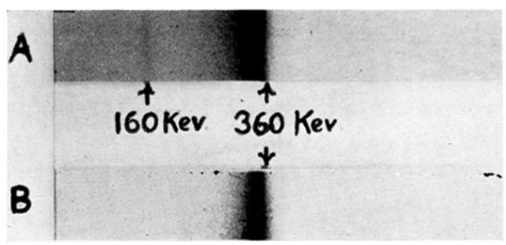

FIG. 3. (A) Electron spectrum of Sr from Rb+H^1. (70 min., 2.75 hr.) (B) Spectrum of Sr bombarded with protons. (80-hr. period).